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## Application of nuclear techniques to environmental plastics research

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### Abstract

Plastic pollution is ubiquitous in aquatic environments and its potential impacts to wildlife and humans present a growing global concern. Despite recent efforts in understanding environmental impacts associated with plastic pollution, considerable uncertainties still exist regarding the true risks of nano- and micro-sized plastics (< 5 mm). The challenges faced in this field largely relate to the methodological and analytical limitations associated with studying plastic debris at low (environmentally relevant) concentrations. The present paper highlights how radiotracing techniques that are commonly applied to trace the fate and behaviour of chemicals and particles in various systems, can contribute towards addressing several important and outstanding questions in environmental plastic pollution research. Specifically, we discuss the use of radiolabeled microplastics and/or chemicals for 1) determining sorption/desorption kinetics of a range of contaminants to different types of plastics under varying conditions, 2) understanding the influence of microplastics on contaminant and nutrient bioaccumulation in aquatic organisms, and 3) assessing biokinetics, biodistribution, trophic transfer and potential biological impacts of microplastic at realistic concentrations. Radiotracer techniques are uniquely suited for this research because of their sensitivity, accuracy and capacity to measure relevant parameters over time. Obtaining precise and timely information on the fate of plastic particles and co-contaminants in wildlife has widespread applications towards effective monitoring programmes and environmental management strategies.

### Keywords

Microplastics; Nuclear applications; Radiotracers; Bioaccumulation

## 1. Introduction

The global proliferation of plastic pollution over the last 60 years, and awareness of its magnitude, has triggered broad public and scientific concern regarding its potential threat to wildlife (Borrelle et al., 2017; Eriksen et al., 2014) and humans through seafood consumption (Barboza et al., 2018; Rochman et al., 2015; Seldenrich, 2015). Research programmes across the world have consequently been directed at understanding and characterising the risks of plastic pollution in fresh- water and marine systems (e.g., GESAMP, 2015; NOAA, 2008; UNEP, 2016). In these studies, the visible impacts of macroplastics (> 250 mm) described are clear (e.g., entanglement, ingestion), but consequences associated with smaller nano- and micro-sized plastics (< 5 mm) are much less obvious. This has led to some controversy on the relative importance of small plastic particles to cause effects in wildlife at environmental concentrations, and a push for improved ecotoxicity research to achieve accurate and reliable risk assessments (Burton, 2017; Connors et al., 2017; Hale, 2018; Koelmans et al., 2017; Kramm et al., 2018).

Recent models estimated that over 5.2 trillion micro-sized plastic particles (0.33–200 mm) weighing 66,140 tonnes are floating in the ocean (Eriksen et al., 2014). As a result, microplastics have been identified in many aquatic organisms, and linked to a range of biological effects (reviewed by Avio et al., 2017; Bouwmeester et al., 2015; Eerkes-Medrano et al., 2015; Ivar Do Sul and Costa, 2014; Wright et al., 2013a). Reported effects include reduced feeding (Besseling et al., 2013; Cole et al., 2015; Wright et al., 2013b), swimming activity (Chen et al., 2017; Gambardella et al., 2017) and assimilation efficiency (Blarer and Burkhardt-Holm, 2016), altered size (Au et al., 2015; Besseling et al., 2013; Redondo-Hasselerharm et al., 2018), impaired reproduction (Au et al., 2015; Sussarellu et al., 2016) and tissue damage (Lei et al., 2018). Several studies have also found that exposure to microplastic particles influenced the accumulation of co-contaminants (Avio et al., 2015; Besseling et al., 2013; Browne et al., 2013; Chua et al., 2014) and present a potential risk for trophic transfer of both plastics and associated contaminants (Au et al., 2017; Carbery et al., 2018; Chae et al., 2018; Farrell and Nelson, 2013; Setälä et al., 2014). Contrary to these studies, several others have found no apparent effects of plastics on a range of organisms (Bruck and Ford, 2018; Santana et al., 2018; Weber et al., 2018), which highlights the need to better understand the discrepancies between studies, including differences in species sensitivity and experimental design.

Despite reported impacts, uncertainties remain regarding the effects associated with nano- and microplastics under ecologically relevant conditions. This primarily stems from the difficulties associated with quantifying low concentrations of small particles and the challenges involved in characterising plastic polymers (Avio et al., 2017; Lenz et al., 2016; Rocha-Santos and Duarte, 2015; Silva et al., 2018). To date, the majority of environmental surveys have focused on particles between 0.3 and 5 mm (Eriksen et al., 2013; Kovač Viršek et al., 2016; Morét-Ferguson et al., 2010), and few studies have considered smaller sized particles because of the difficulties associated with sampling and sorting small particles (Conkle et al., 2018). Most commonly, plastic particles are quantified and characterised by visual assessment, as this is the simplest and cheapest method available (Hidalgo-Ruz et al., 2012). However, this method was found to commonly misidentify plastics for organic particles or vice versa, and consequently inaccurately estimate plastic concentrations when compared to spectroscopic identification (Lenz et al., 2015; Song et al., 2015). Because of these challenges and uncertainties, the majority of laboratory studies investigating the effects of microplastics have used concentrations several orders of magnitude higher than what is typically found in the environment, and likely overstate the effects of plastics under realistic conditions (Lenz et al., 2016).

Recent publications have challenged the initial overstatements of plastic effects reported due to experimental exposures of organisms to unrealistically high concentrations (Burton, 2017; Koelmans et al., 2017; Ogonowski et al., 2018). There is a general consensus in recent reviews that current research needs to better manage and understand the environmental impacts of microplastics (Au et al., 2017; Conkle et al., 2018; Connors et al., 2017; Duis and Coors, 2016; Wagner et al., 2014). Suggested improvements include: (1) establishing standardised methods for sampling, quantifying and characterising nano- and microplastics, (2) increasing environmental relevance in laboratory testing by considering realistic plastic particle concentrations, a range of plastic types and sizes, as well as the influence of weathering, bio-fouling, and abiotic factors to the plastic behaviour, (3) determining the potential role of plastic particles as vectors of contaminants and the risks associated with metals and trace organic compounds sorbed to them, as well as (4) determining the biological

effects of plastic particles at different levels of biological organisation. Considering the methodological challenges and remaining uncertainties surrounding the environmental effects of microplastic and nanoplastic pollution, it seems clear that new technical approaches are required to advance this area of research. In this perspective article, we aim at highlighting the benefits of radiotracer techniques, and describing how these tools can contribute towards advancing environmental plastic pollution research.

## 2. Future perspectives: applications of radiotracer techniques to environmental plastics studies

### 2.1. Radiotracer techniques

Radiotracer techniques consist of measuring the behaviour and fate of radionuclides or labeled compounds within a given system (reviewed by Kratz and Lieser, 2013). This can be achieved using several types of detectors, including scintillation counters, gas-filled detectors and semiconductor detectors that measure the radiation emitted by the tracers. The distribution of radiotracers within a sample can also be visualised using imaging techniques, including autoradiography, positron emission tomography (PET imaging) and single photon emission computed tomography (SPECT). As such, radiodetectors can be used to qualitatively or quantitatively measure tracers on both a spatial and/or temporal scale. These techniques are well recognised as being highly sensitive, accurate, and relatively rapid compared to other analytical methods, and therefore have a broad range of applications in several fields, including life sciences, chemistry and industrial research (Kratz and Lieser, 2013).

Another important advantage of radiotracing methods is the ability, in some cases, to monitor the fate of radiotracers in vivo, in a non-destructive manner. This can be done using in vivo gamma counting, which allows repeated non-invasive measurements of radioactivity to be measured in real time that can be used to determine uptake, assimilation and elimination kinetics in a range of organisms (Reinfelder et al., 1998a). Non-invasive imaging tools, such as PET and SPECT imaging, can also be used to monitor radiotracers in live organisms over time (Decristoforo et al., 2017). In vivo methods not only reduce the processing time but also reduce the biological variability and the number of organisms required compared to experiments where destructive sampling is required to obtain temporal data (e.g., mass spectrometry based analysis of contaminants extracted from tissue samples or whole animals) (Cresswell et al., 2017).

In ecological and ecotoxicity studies, radioactive isotopes of organic and inorganic chemicals are commonly used to assess the fate and transfer of trace elements and compounds between different environmental components (e.g., water, soil, biota), and to quantitatively assess rates of uptake and depuration, assimilation efficiencies, biological half-lives, routes of uptake and biodistribution of chemicals in biota (Cresswell et al., 2015; Danis et al., 2005; Lanctôt et al., 2017; Metian et al., 2009; Wang and Fisher, 1996). Radiotracers can also be used to study biochemical processes, for examples photosynthesis using  $^{14}\text{CO}_2$  (Maleva et al., 2013; Prasad et al., 2011), calcification using  $^{45}\text{Ca}$  (Houlbrèque et al., 2012; Malone and Dodd, 1967) and the metabolism of radiolabeled molecules (Sprinkle et al., 1975; Thomas and White, 1989).

### 2.2. Application to environmental plastics research

Given their high sensitivity and broad applications, laboratory-based radiotracer techniques can provide critical new information on microplastic interactions and biological impacts within the aquatic environment. Specifically, these tools have the potential to help fill important knowledge gaps that include: 1) the sorption and desorption kinetics of trace pollutants to microplastics (Fig. 1a), 2) the evaluation of the biokinetics, biodistribution and potential biological impacts, and trophic transfer of small plastic particles in biota (Fig. 1b), and 3) the influence of microplastics on bioaccumulation and bioavailability of co-contaminants and essential elements to aquatic organisms (Fig. 1c). By efficiently contributing critical information about such fundamental questions, these tools can help address unresolved questions about the ability of microplastics to bioaccumulate (i.e., translocate across epithelial membranes and enter tissues or circulatory system), their role as vectors of chemical transfer, and subsequently, their potential to accumulate through food webs under typical relevant exposure condition.



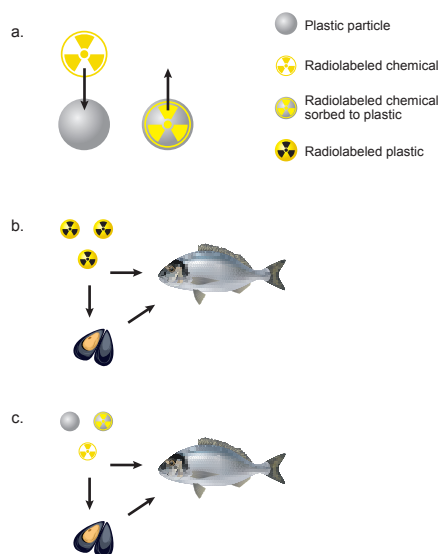


Fig. 1. Schematic representation on how radiotracers can be applied to study (a) the sorption and desorption kinetics of radiolabeled chemicals to plastic particles; (b) the biokinetics, toxicodynamics and potential trophic transfer of microplastics in biota using radiolabeled plastic particles; and (c) the impact of plastics on uptake of co-contaminants and nutrients in aquatic organisms.

#### 2.2.1. Sorption and desorption kinetics

#### 2.2.2.

A growing concern regarding marine plastic pollution is the ability of microplastics to sorb chemicals from the environment and consequently transport and transfer them to aquatic biota via ingestion and/or contact (Bakir et al., 2014a; Besseling et al., 2013; Rochman et al., 2013). Because of their hydrophobic surface, microplastics can readily sorb and concentrate a range of chemical contaminants, including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and trace metals, and there is evidence that some chemicals may preferentially sorb to plastics compared to natural particles in the environment (Beckingham and Ghosh, 2017; Teuten et al., 2007). To accurately assess the role of plastics as vectors in the environment, it is critical to understand the sorption potential and kinetics of a range of chemicals and mixtures to various representative plastic types under realistic (and inherently varying) environmental conditions. This is necessary since the different physical and chemical properties of plastic particles (e.g., polymer, surface area, surface charge, weathering) can affect the sorption of contaminants (Holmes et al., 2012; Teuten et al., 2007; Wang and Wang, 2018), and this can further vary depending on environmental conditions (e.g., salinity, pH) (Bakir et al., 2014b; Turner and Holmes, 2015; Wang et al., 2015, 2018).

It is possible to predict the sorption-desorption kinetics through the use of polymer diffusion models for particles of different sizes (Ahn et al., 2005; Koelmans et al., 2013; Teuten et al., 2009). However, considering the dynamic and complex nature of aquatic systems and the inherent variability in plastic characteristics, it is important to be able to accurately inform and validate these predictive models under controlled conditions. Nuclear tools offer an advantageous complement to this field because radiolabeled chemicals enable sorption and desorption kinetic modelling to be carried out using low concentrations that are within the 'environmentally realistic' range (Reinfelder et al., 1998b), which can be challenging using conventional analytical tools. One of the main advantages of using radiotracers compared to other analytical techniques is their ease of detection with high level precision.

Radiotracers have long been used to study the sorption of organic compounds and trace elements to various matrices (Eichholz et al., 1965; Robertson, 1968; Topp and Smith, 1992) but to date few studies have applied these tools in the context of environmental plastic pollution (Table 1; Bakir et al., 2014a, 2014b, 2012; Napper et al., 2015; Teuten et al., 2007). In fact, only a handful of studies have assessed sorption and desorption kinetics of chemicals to plastics using radiotracers, and the majority have focused on two persistent organic compounds, phenanthrene (Phe) and 4,4'-DDT. Teuten et al. (2007) examined  $^{14}\text{C}$ -Phe ( $0.6\text{--}6.1\ \mu\text{g L}^{-1}$ ) sorption and desorption to three types of

plastics (PE, PVC and PP; 200–250µm), and found that sorption varied greatly on the different plastic types (PE >> PP > PVC). The study also found that sorption was greater and desorption occurred less rapidly for all three plastics compared to two natural sediments tested (Teuten et al., 2007). Bakir et al. (2012) used radiolabeled <sup>3</sup>H-Phe and <sup>14</sup>C-DDT in a mixture to examine the competitive binding of these compounds to PVC and PE particles (also sized at 200–250 µm), and found that DDT had an antagonistic effect on Phe binding to plastics. Similarly, Napper et al. (2015) examined <sup>3</sup>H-Phe and <sup>14</sup>C-DDT sorption to PE microbeads extracted from facial scrubs and found that the different particle types and sizes had varying affinities for persistent organic pollutants (POPs) and that sorption was generally less to the cosmetic-extracted microbeads compared to commercially available PE particles. In another study, Bakir et al. (2014a) examined <sup>14</sup>C-Phe and <sup>14</sup>C-DDT sorption and desorption to PVC and PE particles (200–250 µm) under varying salinity and found that salinity had no significant effect on Phe sorption to plastics but that sorption of DDT was much higher in freshwater compared to seawater. The study also showed that desorption of both Phe and DDT was not influenced by salinity. The same group also compared sorption and desorption of <sup>14</sup>C-labeled Phe, DDT, perfluorooctanoic acid (PFOA) and di-2-ethylhexyl phthalate (DEHP) to PVC and PE (200–250 µm) in seawater and simulated gut conditions under varying pH and temperatures (Bakir et al., 2014a). The study found that under simulated gut conditions the desorption rates of chemicals were much greater compared to seawater, and that pH and temperature also significantly influenced desorption (Bakir et al., 2014a), which highlights the importance of considering environmental realistic conditions for kinetic models. In addition to the few studies on the kinetics of radiolabeled organic compounds, a recent study compared the sorption of radiolabeled Cs and Sr on microplastics, and found that sorption of both elements was generally 2–3 orders of magnitude lower on plastic particles compared to sediment particles (Johansen et al., 2018). This is in contrast with the reported behaviour of organic contaminants where greater sorption to plastic than sediment particles has been found (Beckingham and Ghosh, 2017; Teuten et al., 2007).

Despite recent advances in understanding the influence of environmental parameters on the sorption-desorption of chemicals to plastics (Bakir et al., 2014b; Brennecke et al., 2016; Turner and Holmes, 2015), further research is needed to address several remaining questions in this area (Hartmann et al., 2017; Ziccardi et al., 2016). For example, more research is needed to determine how abiotic physico-chemical or mechanical degradation of microplastics affect sorption/ desorption of chemical contaminants; how the presence of a biofilm formed on virgin or weathered microplastics affect sorption/desorption of chemical contaminants; and how enzymes and different physiological conditions of co-solvents such as gut or fish oil, pH, and temperature can affect desorption of contaminants from plastics contributing to the chemical transfer to the organisms.

**Table 1** Summary of current studies using radiotracers to study environmental effects of microplastics.

Study	Radiotracer(s)	Microplastic (polymer type and size range)	Objective
<i>Sorption and desorption kinetics</i>			
Bakir et al., 2012	<sup>3</sup> H-Phe; <sup>14</sup> C-DDT	PVC, PE; 200–250 µm	Investigate the competitive binding of Phe and DDT in bi-component mixtures to PE and PVC microplastics.
Bakir et al., 2014a	<sup>14</sup> C-Phe; <sup>14</sup> C-DDT	PVC, PE; 200–250 µm	Investigate the effects of salinity on the sorption behaviour of Phe and DDT to PE and PVC microplastics.
Bakir et al., 2014b	<sup>14</sup> C-Phe; <sup>14</sup> C-PFOA; <sup>14</sup> C-DDT; <sup>14</sup> C-DEHP	PVC, PE; 200–250 µm	Compare the sorption behaviour of Phe, DDT, PFOA and DEHP to PE and PVC in seawater and under simulated gut conditions.
Napper et al., 2015	<sup>3</sup> H-Phe; <sup>14</sup> C-DDT	PE; 164–327 µm	Compare the sorption behaviour of Phe and DDT to PE microbeads extracted from cosmetics.
Teuten et al., 2007	<sup>14</sup> C-Phe	PE, PVC, PP; 200–250 µm	Compare the sorption behaviour of Phe to different type of microplastics and natural particles.
Johansen et al., 2018	<sup>134</sup> Cs; <sup>85</sup> Sr	PE; 100 µm	Compare the sorption behaviour of Cs and Sr to PE microplastics with biofilm from different environments.
<i>Biokinetics of plastic particles</i>			
NA			
<i>Role of plastics as vectors of contaminant transfers</i>			
Khan et al., 2015	<sup>110m</sup> Ag	PE; 10–106 µm	Investigate the effect of PE microplastics on the uptake and localization of Ag in zebrafish ( <i>Danio rerio</i> ).
Khan et al., 2017	<sup>110m</sup> Ag	PE; 10–106 µm	Investigate the effect of PE microplastics on intestinal uptake of Ag in rainbow trout ( <i>Oncorhynchus mykiss</i> ).
Ma et al., 2016	<sup>14</sup> C-Phe	PS; 50 nm, 500 nm, 5 µm, 10 µm, 15 µm	Investigate the effect of nano and micro-sized plastics on Phe uptake and toxicity in <i>Daphnia magna</i> .
Frydkjaer et al., 2017	<sup>14</sup> C-Phe	PE; 10–75 µm	Investigate the effect of PE microplastics on Phe uptake and toxicity in <i>Daphnia magna</i> compared to different plankton species.
<i>Tissue distribution of plastics and associated contaminants</i>			
NA			

Phe, phenanthrene; DDT, 4,4'-dichlorodiphenyltrichloroethane.

PVC, polyvinyl chloride; PE, polyethylene; PP, polypropylene; PS, Polystyrene.

NA, not available.

### 2.2.2. Biokinetics of plastic particles

To understand the risks associated with microplastic pollution and their role in the transfer of contaminants, it is imperative that we also understand the physical effects of plastic particles themselves, including their accumulation, translocation and trophic transfer within the environment. To study the accumulation of plastic particles by biota most studies perform whole body or tissue-specific measurements by digesting tissues and separating particles by filtration (Catarino et al., 2017; Cole et al., 2014) or use tissue sectioning and histological assessment (Avio et al., 2015; Sussarellu et al., 2016), both followed by visual and/or spectroscopy confirmation. In laboratory studies, fluorescent microbeads are also commonly used for their practicality (i.e., ease of counting by flow-cytometry and visualisation using fluorescent microscope) (Cole et al., 2013; Lu et al., 2016). Nevertheless, these techniques are generally limiting in quantifying low concentrations of small particles, and require large sample sizes to obtain temporal data.

The limitations encountered using traditional techniques could be resolved using nuclear methodologies by creating radioactive plastic particles that can be traced using conventional radiotracing tools. This approach has historically been used for medical and physiological studies (Reinhardt et al., 2001; Rudolph and Heymann, 1967; Wolen et al., 1984; Yipintsoi et al., 1973), but to the best of our knowledge, it has never been applied in the context of microplastic impact assessment. Most commonly, radiolabeled microspheres have been synthesized from carbonised plastic (e.g., polystyrene) embedded with trace amounts of radionuclides (e.g.,  $^{51}\text{Cr}$ ,  $^{125}\text{I}$ ,  $^{85}\text{Sr}$ ,  $^{46}\text{Sc}$ ; Fig. 2a). Though this method has proven very efficient for acute studies, radiolabeled microspheres have been reported to be less efficient in chronic test due to leaching of radioisotopes from the spheres (Van Oosterhout et al., 1998; Glenny et al., 1993). In order to circumvent the issue to leaching from microspheres embedded in radioisotopes, radiolabeled plastic particles could be synthesised by either encapsulating a nano-radionuclide (e.g.,  $^{110\text{m}}\text{Ag}$ ) core with plastic polymer (Fig. 2b) or coating a nanoparticle with plastic polymer and inducing radioactivity by neutron activation of the element forming the core in a nuclear reactor (Fig. 2c). By containing the isotope within the plastic sphere any leaching should theoretically be avoided. We are currently developing the methods for producing radiolabeled microspheres for use in environmental plastics research.

This type of radiolabeled plastic particles would be extremely well suited to study the biokinetics, biodistribution, and trophic transfer of the particles themselves. For example, this novel approach could provide accurate assessments of gut retention times, particle assimilation (i.e., if they can cross epithelial membranes), and the relative importance of different uptake routes (e.g., digestive tract, gills, skin) across a range of species. These questions are important to consider, not only to evaluate the physical impacts of microplastics, but also because the biokinetic is an important factor determining the bioaccumulation potential of co-contaminants. Additionally, given the sensitivity of the radio-detectors, radiolabeled plastic particles would enable very low environmental concentrations to be assessed (presumably individual particles could be detected). This has been the case for radiolabeled nanoparticles, that have similarly been used to understand the environmental fate and bioaccumulation of metal-containing engineered nanoparticles (reviewed by Yin et al. (2017), and for which radio-detection has enabled environmental concentrations to be studied (Al-Sid-Cheikh et al., 2013).

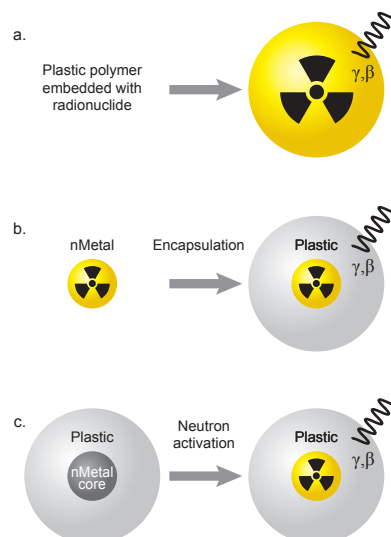


Fig. 2. Schematic representation of different types of radiolabeled plastic microspheres comprised of (a) plastic polymer embedded with a specific radionuclide, (b) radiolabeled nano-metal particle encapsulated with plastic polymer or (c) a nano-metal core (e.g., nAg) coated with a plastic polymer that is neutron activated to induce radioactivity.

### 2.2.3. Role of plastics as vectors of contaminant transfers

Plastics can efficiently sorb various types of toxic compounds, including POPs and trace metals. As such, they may act as both sources of, and long-range vectors for, chemical pollutants in the marine environment (Alimi et al., 2018; Avio et al., 2015; Brennecke et al., 2016). Chemicals sorbed to plastic particles can enter trophic networks through different pathways, potentially enhancing bioaccumulation and/or biomagnification efficiencies. Aquatic organisms, ranging from invertebrates to aquatic mammals, have been found to ingest plastic particles in their natural environment (Bravo Rebolledo et al., 2013; Desforges et al., 2015; Lusher et al., 2015; Steer et al., 2017). It has further been suggested that contaminants sorbed to microplastics can subsequently be transferred to the animals that ingest them (reviewed by Hartmann et al., 2017; Koelmans et al., 2016; Ziccardi et al., 2016). For example, it has been reported that PS microplastics increase PCB bioaccumulation in lugworms (*Arenicola marina*) when co-exposed in sediment (Besseling et al., 2013; Koelmans et al., 2013). Nevertheless, it has also been hypothesized that the presence of microplastics could reduce co-contaminant accumulation or have no influence (Koelmans et al., 2016).

Uncertainties regarding the importance of plastics as sources of contaminant transfer under natural conditions must be addressed (Koelmans et al., 2016). A key limiting factor in effectively addressing questions of plastic-mediated chemical transfer is the limit of detection of common analytical tools. Effectively extracting, isolating and quantifying chemical contaminants using traditional mass spectrometry based analytical technologies poses difficulties and it has thus far proven difficult to explore this question using these techniques. Highly sensitive nuclear and isotopic techniques could therefore meaningfully contribute towards determining the role of microplastics as vectors of co-contaminant bioaccumulation/bioavailability. These tools can readily be applied to study whether the presence of plastic in the environment enhances uptake/loss of co-contaminants or essential elements in aquatic organisms, and how transfer via plastics compares to other types of naturally occurring particles. This can be achieved by exposing organisms to radiolabeled chemicals (e.g.,  $^{14}\text{C}$ -organic pollutants or trace element radioisotopes) with or without plastic (or other) particles in water, sediment and/or diet.

To date, only a few studies have used radiotracers to investigate chemical transfer to aquatic organisms by microplastics (Table 1). Khan et al. (2015) utilised radiolabeled silver ( $^{110}\text{mAg}$ ) to assess whether the uptake and biodistribution of Ag ( $1\text{ }\mu\text{g L}^{-1}$ ) was influenced by PE microbeads (10, 100, 1000 beads  $\text{mL}^{-1}$ ; 10–106  $\mu\text{m}$ ) in zebrafish (*Danio rerio*). Though the study found no direct

impact of microplastics on Ag bioaccumulation, they suggested that microplastics can influence the bioavailability and route of uptake of contaminants. A more recent study by the group also assessed the impacts of microplastics on intestinal uptake of  $^{110m}\text{Ag}$  using intestine gut sac preparations (from rainbow trout, *Oncorhynchus mykiss*) (Khan et al., 2017). They reported that microplastics have no significant influence on trans-epithelial transport of Ag since the dissociation of sorbed Ag occurs prior to reaching the site of uptake in the digestive tract ( $> 98\%$  desorption of Ag from microplastics at pH 2.2 and 4.1), but concluded that microplastics can indeed introduce labile contaminants into the intestine (Khan et al., 2017). In another study, Ma et al. (2016) used radiolabeled  $^{14}\text{C}$ -Phe ( $0.1 \text{ mg L}^{-1}$ ) to investigate the effect of nano- and micro-sized PS particles ( $50 \text{ nm}$  and  $10 \text{ }\mu\text{m}$ ) co-exposure on Phe uptake by the cladoceran *Daphnia magna*. The study concluded that only the nano-sized particles ( $50\text{nm}$ ) significantly enhanced bioaccumulation of Phe throughout the 14-day experiment and an additive effect was observed both due to Phe toxicity and due to plastic nanoparticles entering the organism body. Similarly, Frydkjær et al. (2017) tested the influence of PE microplastics on  $^{14}\text{C}$ -Phe ( $12 \text{ }\mu\text{g L}^{-1}$ ) uptake by *D. magna*, but also investigated differences between regular shaped beads ( $10\text{--}106\mu\text{m}$ ) and irregular shaped fragments ( $10\text{--}75 \text{ }\mu\text{m}$ ). The study found that *D. magna* ingested both regular and irregular shaped plastic particles but the egestion rate of irregular fragments was much slower than regular shaped beads, highlighting the importance of considering natural plastic weathering in future studies (Frydkjær et al., 2017). In addition to these observations, the study compared the affinity of  $^{14}\text{C}$ -Phe to PE plastics to different plankton organisms (yeast, bacterium and mixed plankton) and found that sorption was 8–35 times less efficient on plastic fragments compared to plankton, making these organisms much more important vectors of Phe transfer than plastics given their relative prevalence in the environment. Though these few studies demonstrate the use of radiotracers in environmental microplastic research, the full potential of nuclear applications are far from being fulfilled in this field.

#### 2.2.4. Tissue distribution of plastics and associated contaminants

Biokinetic studies in aquatic organisms can be complemented with nuclear imaging techniques to obtain information on the distribution patterns of radiotracers, plastic particles or non-labeled elements of interest in whole animals, tissues, cells, or subcellular fractions. Most commonly, nuclear imaging tools such as autoradiography, positron emission tomography (PET imaging), and single photon emission computed tomography (SPECT) rely on radiotracers to obtain 2- or 3- dimensional images of their distribution in vivo or post-mortem (Decristoforo et al., 2017). In addition, other tools such as X-ray imaging (e.g., X-ray fluorescence microscopy (XFM), particle induced X-ray emission (PIXE)), can also be used to obtain high-resolution images of elemental patterns and do not necessitate the use of radiotracers (Hare et al., 2015). To date, the visualisation of plastic particles within organisms is mostly done using microscopic analysis of fluorescent microspheres (Batel et al., 2018; Cole et al., 2013; Dawson et al., 2018; Setälä et al., 2014), and though this non-nuclear technique has many advantages, it can prove difficult to detect low concentrations of plastics. Nuclear imaging tools have the advantage of being highly sensitive and accurate; thereby facilitating the visualisation of trace concentrations of particles or tracers. In addition to being used for biodistribution assessments, imaging tools can also serve to measure morphological impacts of plastics at both the tissue and organism level. In the context of plastics research, imaging tools could be applied to study if nano- and micro-sized particles are transported across cellular membranes, if plastic particles influence the route of uptake and internal distribution

of associated chemicals or if ingestion or contact causes tissue damage. Nevertheless, in spite of their broad applicability and sensitivity nuclear tools have rarely been applied towards plastics research.

### 3. Challenges and limitations

Despite the many advantages of radiotracer techniques, these specialised techniques present a unique set of technical challenges that are important to consider. Most notably are:

- The need for specialised equipment and facilities to undertake experimental work with radioactive substances.
- Safety and technical consideration of handling of radioactive materials, including radiological protection and regulations, permissible limits of residual activity, waste disposal, etc.

- Limitations in outsourcing analyses of radioactive samples, which may be difficult or impossible if the equipment required is not available in-house.

Further to these logistical limitations, the use of radiotracers also involves several specific considerations (Kratz and Lieser, 2013; Ruth, 2009). Specifically, radiotracers require that the radiation emitted has no physical or biological impact on the system under study. Tracers must also behave identically to the traced substance (e.g., activated plastic particles must behave identically to normal plastic particles therefore thorough characterisation of particles is necessary). The half-life of radionuclides must also be sufficiently long for experimental needs, and thus limits the choice of elements or molecules. Radiotracing methods also require specific quality assurance and quality control measures, including appropriate standards (e.g., geometric standards) and calibrations, to ensure optimal data quality (Cresswell et al., 2017).

In addition to the methodological considerations specific to nuclear tools, applying these methods to assess the impacts of plastic pollution will also involve some of the many challenges encountered in environmental plastic research (Au et al., 2017; Burton, 2017; Connors et al., 2017; Duis and Coors, 2016). These include, but are not limited to, accurately quantifying low concentrations of plastic particles, efficiently characterizing particles (type, size, shape, density, degree of weathering, etc.) and inter-particle interactions (aggregations), as well as maintaining environmental relevance in a controlled setting.

#### 4. Conclusions

Radiotracer techniques have been used in research for over a century (Hevesy and Paneth, 1913) and their benefits are well demonstrated and recognised in medical, industrial, biological and environmental research (Lappin, 2015; Ruth, 2009). Nevertheless, these techniques are greatly underutilised in non-medical fields, particularly in environmental and ecotoxicological research, where they have proven to be extremely valuable (Cresswell et al., 2017, 2015; Lanctôt et al., 2017; Metian et al., 2009; Pouil et al., 2017). The present perspective endeavoured to highlight the usefulness of nuclear applications to significantly complement research in the emerging field of environmental plastic pollution. Given the current need to better understand the impacts of plastic pollution on the environment and human health, and technical challenges in the field, it is critical that we start exploring options beyond standard risk assessment approaches. Radiotracing techniques are some of the most sensitive and accurate methods to trace the movement of chemicals and particles *in vivo*. As such, we believe these tools are well suited to contribute towards resolving central knowledge gaps in environmental plastics research, including their biokinetics and toxicodynamics under environmental conditions, their role as vectors of contaminant transfer, and their potential to accumulate and biotransfer within aquatic food chains. Acquiring this type of precise and timely information on the potential environmental impacts associated with microplastic pollution has widespread applications towards effective monitoring programmes and environmental management strategies.

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